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Description

This invention relates to thermoplastic films, sheets and laminates and coextruded materials formed therefrom, and films and sheets formed from blends of an olefin polymer composition with other olefin polymer materials.

In many film applications, such as packaging of foodstuffs, chemical and hazardous materials and in medical applications, the industry requires films having certain properties. In the packaging of foodstuffs, for example, the films must have high puncture resistance, high clarity and gloss, and reduced permeability to gases and/or vapors. The films used to manufacture containers for chemicals and hazardous waste materials must have a high puncture resistance, high elongation strength, high tear resistance and chemical resistance. Films used in medical applications, such as blood bags, must have a high puncture resistance, low modulus, high tear resistance and auto-clavability.

Films made from ethylene polymers, e.g., HDPE and LLDPE, and propylene polymers, such as crystalline homopolymers of propylene and random copolymers of propylene and ethylene, do not provide such a combination of desirable properties.

Attempts have been made to overcome the shortcomings of these polymers by preparing heterophasic mixtures of crystalline propylene polymers and 8 to 25% of an elastomeric propylene-ethylene copolymer by sequential polymerization in the presence of a stereospecific Ziegler-Natta type catalyst. However, films of such heterophasic compositions are subject to the formation of fisheyes, inadequate tear strength or the formation of rough surfaces.

Hence, there is a need for a polymer material which has a lower flexural modulus, high clarity, good tear strength, as well as all of the other desirable properties.

All parts and percentages used in this application are by weight unless otherwise specified. Ambient or room temperature is approximately 25°C.

This invention provides a thermoplastic film having the desired properties comprising a heterophasic olefin polymer composition which is comprised of

(a) from about 10 to 50 parts of a propylene homopolymer having an isotactic index greater than 80, or a copolymer selected from the group consisting of (i) propylene and ethylene, (ii) propylene, ethylene and a $\text{CH}_2 = \text{CHR}$ alpha-olefin, where R is a C_{2-8} straight or branched alkyl, and (iii) propylene and an alpha-olefin as defined in (a) (ii), said copolymer containing over 80% propylene and having an isotactic index greater than 80;

(b) from about 5 to 20 parts of a semi-crystalline, essentially linear copolymer fraction having a crystallinity of about 20 to 60% wherein the copolymer is selected from the group consisting of (i) ethylene and propylene containing over 55% ethylene, (ii) ethylene, propylene, and an alpha-olefin as defined in (a) (ii) containing from 1 to 10% of the alpha-olefin and over 55% of both ethylene and alpha-olefin, and (iii) ethylene and an alpha-olefin as defined in (a) (ii) containing over 55% of said alpha-olefin, which copolymer is insoluble in xylene at room or ambient temperature; and

(c) from about 40 to 80 parts of a copolymer fraction is selected from the group consisting of a copolymer of (i) ethylene and propylene wherein the copolymer contains from 20% to less than 40% ethylene and, (ii) ethylene, propylene, and an alpha-olefin as defined in (a) (ii) wherein the alpha-olefin is present in an amount of 1 to 10% and the amount of ethylene and alpha-olefin present is from 20% to less than 40%, and (iii) ethylene and an alpha-olefin as defined in (a) (ii) containing from 20% to less than 40% of the alpha-olefin, and optionally with 0.5 to 10% of a diene, said copolymer fraction being soluble in xylene at ambient temperature, and having an intrinsic viscosity of from 1.5 to 4.0 dl/g;

with the total of the (b) and (c) fractions, based on the total olefin polymer composition, being from about 50% to 90%, and the weight ratio of (b)/(c) being less than 0.4.

In another embodiment this invention provides films comprising a layer of the heterophasic olefin polymer composition applied to at least one surface of a thermoplastic film material or a metallic substrate.

In yet another embodiment films from blends of the olefin polymer composition and a thermoplastic material are provided.

Component (a) is preferably present in an amount from 10 to 40 parts, most preferably from 20 to 35 parts. When (a) is a propylene homopolymer, the isotactic index is preferably from about 85 to 98. When (a) is a copolymer, the amount of propylene in the copolymer is preferably from about 90 to 99%.

Component (b) is preferably present in an amount from 7 to 15 parts. Typically the crystallinity is about 20 to 60% by differential scanning calorimetry (DSC). Generally, the ethylene or said alpha-olefin content or the combination of ethylene and said alpha-olefin when both are used is over 55% up to 98%, preferably from 80 to 95%.

Component (c) is preferably present in an amount from 50 to 70 parts. The ethylene or said alpha-olefin content or ethylene and said alpha-olefin content of component (c) is preferably from 20 to 38%, most preferably from 25 to 38%. When component (c) is a terpolymer, the said alpha-olefin is typically present in an amount from 1 to 10%, preferably 1 to 5%. The preferred intrinsic viscosity of is from 1.7 to 3 dl/g.

5 The total amount of (b) and (c), based on the total olefin polymer composition is preferably from 65 to 80% and the weight ratio of (b)/(c) is preferably from 0.1 to about 0.3.

The total amount of ethylene units or said alpha-olefin units, or of ethylene and said alpha-olefin units when both are present, in the olefin polymer composition is from about 15% to about 35%.

10 The compositions have at least one melt peak, determined by DSC, present at temperatures higher than 120 °C, and at least one peak, relative to the vitreous transition, present at temperatures from -10 °C and -35 °C.

15 In addition, these compositions have a flexural modulus of less than 150 MPa, generally from 20 and 100 MPa; a tensile strength at yield of from 10 to 20 MPa, elongation at break over 400%; a tension set, at 75% strain, from 20% to 50%; a Shore D hardness from 20 and 35; and do not break (no brittle impact failure) when an IZOD impact test is conducted at -50 °C.

Preferably the haze values are less than 40%, most preferably less than 35%.

Copolymer and terpolymers of propylene and ethylene or an alpha-olefin or of propylene, ethylene and an alpha-olefin are preferred as component (a), and copolymers of propylene with ethylene or an alpha-olefin are most preferred as component (a) since they provide the compositions with higher clarity.

20 Suitable alpha-olefins of the formula $\text{CH}_2 = \text{CHR}$ include butene-1, pentene-1, 4-methylpentene-1, hexene-1, and octene-1. When used to prepare component (a) they are present in such quantities that the isotactic index of the resulting polymer is not less than 80%.

25 When a diene is present during the preparation of components (b) and (c), it is typically a butadiene, 1,4-hexadiene, 1,5-hexadiene, ethylidene norbornene diene monomer and is typically present in amount from 0.5 to 10%, preferably 1 to 5%.

30 The compositions can be prepared with a polymerization process comprising at least two stages, where in the first stage the propylene or propylene and ethylene or said alpha-olefin or propylene, ethylene or said alpha-olefin are polymerized to form component (a), and in the following stages the mixtures ethylene and propylene or said alpha-olefin or ethylene, propylene and said alpha-olefin, and optionally a diene, are polymerized to form components (b) and (c).

35 The polymerization can be conducted in liquid phase, gas phase, or liquid-gas phase using separate reactors, all of which can be done either by batch or continuously. For example, it is possible to carry out the polymerization of component (a) using liquid propylene as diluent, and the polymerization of components (b) and (c) in gas phase, without intermediate stages except for the partial degassing of the propylene. This is the preferred method.

The polymerization reactions are carried out in an inert atmosphere in the presence of an inert hydrocarbon solvent or of a liquid or gaseous monomer.

Suitable inert hydrocarbon solvents include saturated hydrocarbons, such as propane, butane, hexane and heptane.

40 Hydrogen can be added as needed as a chain transfer agent for control of the molecular weight.

The reaction temperature in the polymerization of component (a) and for the polymerization of components (b) and (c), can be the same or different, and is generally from 40 °C to 90 °C, preferably 50 to 80 °C for the polymerization of component (a), and 40 to 65 °C for the polymerization of components (b) and (c).

45 The pressure of the polymerization of component (a), if carried out in liquid monomer, is the one which competes with the vapor pressure of the liquid propylene at the operating temperature used, eventually modified by the vapor pressure of the small quantity of inert diluent used to feed the catalyst mixture, and the overpressure of optional monomers and the hydrogen used as molecular weight regulator.

50 The pressure of the polymerization of components (b) and (c), if done in gas phase, can be from 5 to 30 atm. The residence times relative to the two stages depend on the desired ratio between fraction (a) and (b) + (c), and are usually from 15 min. to 8 hours.

55 The catalyst used in the polymerization comprises the reaction product of 1) a solid component containing a halogen-containing titanium compound and an electron-donor compound (internal donor) supported on an activated magnesium chloride, 2) a non-halogen containing Al-trialkyl compound and 3) an electron-donor compound (external donor).

Suitable titanium compounds include those with at least one Ti-halogen bond, such as halides and alkoxy halides of titanium.

In order to obtain these olefin polymer compositions in the form of flowable spherical particles having a high bulk density, the solid catalyst component must have a) a surface area smaller than 100 m²/g, preferably between 50 and 80 m²/g, b) a porosity from 0.25 to 0.4 cc/g. and c) an X-ray spectrum, where the magnesium chloride reflections appear, showing the presence of a halo between the angles 2 ϑ of 33.5° and 35° and by the absence of the reflection at 2 ϑ of 14.95°. The symbol ϑ = Bragg angle.

The solid catalyst component is prepared by forming an adduct of magnesium dichloride and an alcohol, such as ethanol, propanol, butanol and 2-ethylhexanol, containing generally 3 moles of alcohol per mole of MgCl₂, emulsifying the adduct, cooling the emulsion quickly to cause the adduct to solidify into spherical particles, and partially dealcoholating the particulate adduct by gradually increasing the temperature from 50°C to 130°C for a period of time sufficient to reduce the alcohol content from 3 moles to 1-1.5 moles per mole of MgCl₂. The partially dealcoholated adduct is then suspended in TiCl₄ at 0°C, such that the concentration of adduct to TiCl₄ is 40-50 g/l TiCl₄. The mixture is then heated to a temperature of 80°C to 135°C for a period of about 1-2 hr. When the temperature reaches 40°C, sufficient electron donor is added so that the desired molar ratio of Mg to electron donor is obtained.

An electron-donor compound selected preferably among the alkyl, cycloalkyl, and aryl phthalates, such as for example diisobutyl, di-n-butyl, and di-n-octyl phthalate, is added to the TiCl₄.

When the heat treatment period has ended, the excess hot TiCl₄ is separated by filtration or sedimentation, and the treatment with TiCl₄ is repeated one or more times. The solid is then washed with a suitable inert hydrocarbon compound, such as hexane or heptane, and dried.

The solid catalyst component typically has the following characteristics:

Surface area:	less than 100 m ² /g, preferably between 50 and 80 m ² /g
Porosity:	0.25 - 0.4 cc/g
Pore volume distribution:	50% of the pores have a radius greater than 100 angströms.
X-ray spectrum:	where the Mg chloride reflections appear, showing a halo with maximum intensity between angles of 2 ϑ of 33.5° and 35°, and where the reflection at 2 ϑ of 14.95° is absent.

The catalyst is obtained by mixing the solid catalyst component with a trialkyl aluminum compound, preferably triethyl aluminum and triisobutyl aluminum, and an electron-donor compound.

Various electron donor compounds are known in the art. The preferred electron donor compounds are those silane compounds having the formula R'R"Si(OR)₂ where R' and R" may be the same or different and are C₁₋₈ normal or branched alkyl, C₅₋₁₈ cycloalkyl, or C₆₋₁₈ aryl radicals, and R is a C₁₋₄ alkyl radical.

Typical silane compounds which may be used include diphenyldimethoxysilane, dicyclohexyldimethoxysilane, methyl-t-butylidemethoxysilane, diisopropyldimethoxysilane, dicyclopentyldimethoxysilane, cyclohexylmethyldimethoxysilane and phenyltrimethoxysilane.

The Al/Ti ratio is typically between 10 and 200 and the Al/silane molar ratio between 1/1 and 1/100.

The catalysts may be precontacted with small quantities of olefin monomer (prepolymerization), maintaining the catalyst in suspension in a hydrocarbon solvent and polymerizing at a temperature from room temperature to 60°C for a time sufficient to produce a quantity of polymer from 0.5 to 3 times the weight of the catalyst.

This prepolymerization also can be done in liquid or gaseous monomer to produce, in this case, a quantity of polymer up to 1000 times the catalyst weight.

The content and amount of catalyst residue in the thermoplastic olefin polymers of this invention is sufficiently small so as to make the removal of catalyst residue, typically referred to as deashing, unnecessary.

The thermoplastic olefin polymers prepared with the aforementioned catalyst are in spheroidal particle form, and the particles have a diameter from 0.5 to 7 mm.

Unless otherwise specified, the following analytical methods are used to characterize the supported catalyst component, the heterophasic olefin polymer compositions, films prepared therefrom and comparative film materials.

<u>Properties</u>	<u>Method</u>
Melt Flow Rate, g/10 min.	ASTM-D 1238, condition L
5 Ethylene, wt %	I. R. Spectroscopy
Intrinsic viscosity	Determined in tetrahydro-naphthalene at 135°C
10 Xylene solubles, wt %	See description below.
15 Flexural modulus at 23°C and Vitreous transition temperature	Using a device for dynamic-mechanical measurements of DMTA of Polymer Laboratories at a frequency measure of 1 Hz and a scanning temperature of 2°C/min. A sample plaque (40x10x2 mm) of the polymer to be analyzed is cut from a pressure molded sheet prepared with a Carver press at 200°C with 10 tons of pressure for 10 minutes and then cooling the sheet at 15°C/min.
20	
25	
Notched IZOD impact	ASTM-D 256
30 Haze	ASTM-D 1003
35	
40	
45	
50	
55	

<u>Properties</u>	<u>Method</u>
Tension set at 75%	ASTM-D 412
5 Tensile Strength at yield and at break	ASTM-D 638
10 Elongation at yield and at break	ASTM-D 638
Surface area	B.E.T.
15 Porosity	B.E.T.
Bulk density	DIN-53194
20 Elemendorf tear	ASTM-D 1922-78
Trouser tear	ASTM-D 1938-85
25 Gloss	ASTM-D 523
Coefficient of friction	ASTM-D 1894
Moisture vapor transmission	ASTM-E 96
30 Dart impact strength	ASTM D 4272-83

30 Unless otherwise specified, the samples of the olefin polymer composition to be subjected to the various physical-mechanical analyses are molded by use of a Negri & Bossi 90 injection press, after stabilizing the material with 0.1% Irganox 1010 tetrakis[methylene 3,5-di-tert-butyl-4-hydroxyhydrocinnamate] methane and 0.1% BHT (2,6-di-tert-butyl-p-cresol), and pelletizing it with a single screw Bandera extruder (cylinder diameter 30 mm) at 210°C. The analytical conditions are as follows:

35 temperature of the melt 250°C
temperature of the mold 60°C
injection time 20 sec
cooling time 25 sec.

The samples to be subjected to the Haze analysis are molded into 75x75x1 mm plaques using a GBF F 40 235/90 injection press under the following conditions:

temperature of the melt 260°C
temperature of the mold 40°C
injection time 20 sec
cooling time 10 sec.

45 The samples of the film materials were 1 mil in thickness and were cut from the film sheet in the size provided in the particular ASTM test method being used.

The weight percentage of the sum of the (b) and (c) fractions, indicated by % (b) + (c), is calculated by determining the weight of the mixture fed during the second stage, and comparing it with the weight of the final product.

50 The weight percentage (%) of the (a), (b), and (c) fractions described herein are determined as follows:

$$\% (a) = 100\% - [(b) + (c)]$$

$$\% (c) = S_f P_a S_a$$

55 where S_f and S_a are the percentage by weight of the portion soluble in xylene of the final product and the polypropylene fraction (a), respectively; P_a is the weight ratio between said fraction and the final product.

$$\% (b) = 100 - \% (a) - \% (c)$$

The percentage by weight of ethylene or said alpha-olefin or ethylene and said alpha-olefin contained in copolymer fraction (c) soluble in xylene is calculated using the following formula:

5

$$\text{wt.\% ethylene and/or said alpha-olefin in fraction (c)} = \frac{(C_f - C_a) \cdot X}{1 - X}$$

10

where:

C_f = % ethylene and/or said alpha-olefin in the xylene solubles of the final product;

C_a = % ethylene and/or said alpha-olefin in the xylene solubles of fraction (a);

X = $S_z \cdot P_a/S_f$

15

The intrinsic viscosity of fraction (c), (I.V._(c)), is calculated using the following formula:

$$(I.V._{(c)}) = (I.V._{S_f} - I.V._{(a)}) \cdot X / (1 - X)$$

20

where $I.V._{S_f}$ is the intrinsic viscosity of the xylene soluble fraction of the final composition and $I.V._{(a)}$ is the intrinsic viscosity of the xylene soluble portion of fraction (a).

25

The weight percent of olefin polymer composition soluble in xylene at room temperature is determined by dissolving 2.5 g of the polymer in 250 ml of xylene in a vessel equipped with a stirrer which is heated at 135 °C with agitation for 20 minutes. The solution is cooled to 25 °C while continuing the agitation, and then left to stand without agitation for 30 minutes so that the solids can settle. The solids are filtered with filter paper, the remaining solution is evaporated by treating it with a nitrogen stream, and the solid residue is vacuum dried at 80 °C until a constant weight is reached. The percent by weight of polymer insoluble in xylene at room temperature is the isotactic index of the polymer. The value obtained in this manner corresponds substantially to the isotactic index determined via extraction with boiling n-heptane, which by definition constitutes the isotactic index of the polymer.

30

Further embodiments of the invention can be seen from the dependant claims.

Examples illustrative of the heterophasic olefin polymer composition, physical properties thereof, a process for preparing same, a film based on said olefin polymer composition and a method of preparing said film are set forth below.

35

Solid Catalyst Component

A) Preparation of MgCl₂/Alcohol Adduct

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Under an inert atmosphere, 28.4 g anhydrous MgCl₂, 49.5 g of an anhydrous ethanol, 100 ml of ROL OB/30 vaseline oil and 100 ml of silicone oil having a viscosity of 350 cs are introduced into a reaction vessel equipped with a stirrer and heated at 120 °C with an oil bath and stirred until the MgCl₂ is dissolved. The hot reaction mixture is then transferred under inert atmosphere to a 1500 ml vessel equipped with an Ultra Turrax T-45 N stirrer and a heating jacket and containing 150 ml of vaseline oil and 150 ml of silicone oil. The temperature is maintained at 120 °C with stirring for 3 minutes at 3,000 rpm. The mixture is then discharged into a 2 liter vessel equipped with a stirrer containing 1,000 ml of anhydrous n-heptane cooled at 0 °C with a dry ice/isopar bath and stirred at a tip speed of 6 m/sec for about 20 minutes while maintaining the temperature at 0 °C. The adduct particles thus formed are recovered by filtering, are washed 3 times at room temperature with 500 ml aliquots of anhydrous hexane and gradually heated by increasing the temperature from 50 °C to 100 °C under nitrogen for a period of time sufficient to reduce the alcohol content from 3 moles to 1.5 moles per mole of MgCl₂. The adduct has a surface area of 9.1 m²/g and a bulk density of 0.564 g/cc.

B) Solid Catalyst Component Preparation

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The adduct (25 g) is transferred under nitrogen into a reaction vessel equipped with a stirrer and containing 625 ml of TiCl₄ at 0 °C under agitation. It is then heated to 100 °C in 1 hr. When the temperature reaches 40 °C, diisobutylphthalate is added in an amount such that the molar ratio of Mg to diisobutylphthalate is 8. The contents of the vessel are heated at 100 °C for 2 hours with agitation, the agitation is

stopped and the solids are allowed to settle. The hot liquid is removed by siphon. 550 ml of TiCl₄ is added to the solids in the vessel and the mixture heated at 120 °C for 1 hr. with agitation. The agitation is stopped and the solids are allowed to settle. The hot liquid is then removed by siphon. The solids are washed 6 times at 60 °C with 200 ml aliquots of anhydrous hexane, and then 3 times at room temperature. The solids, after being vacuum dried, have a porosity of 0.261 cc/g, a surface area of 66.5 m²/g and a bulk density of 0.44 g/cc.

Examples 1-3

These examples illustrate the heterophasic olefin polymer composition and a method for preparing the polymers.

General Operating Conditions

The polymerization runs are conducted under nitrogen in a 22 liter stainless steel autoclave equipped with a helicoid magnetic stirrer and operated at about 90 rpm. All temperatures, pressures and concentrations of olefin monomers and hydrogen, when present, are constant unless otherwise indicated. The concentration of hydrogen and of the relative monomers is analyzed continuously in gas phase with a process gas chromatograph and fed in order to maintain constant the desired concentration of same.

The polymerization is a batch process conducted in two stages. The first stage comprising the polymerization of the relevant monomer or monomers in liquid propylene and the second stage the copolymerization of ethylene and propylene in gas phase.

In the first stage, the following ingredients in the order in which they are listed are fed into the autoclave at 20 °C over a period of about 10 minutes: 16 l of liquid propylene, appropriate quantities of ethylene and hydrogen, and the catalyst system consisting of 1) the solid catalyst component (about 0.15g) prepared as described above, and 2) a mixture of 75 ml of triethyl aluminum (TEAL) at a 10% concentration in hexane and an appropriate quantity of cyclohexylmethyldimethoxysilane (CMMS) electron donor such that the Al/CMMS molar ratio is 7.5. The catalyst system is pressure fed into the autoclave with propylene.

The temperature is brought to the desired level in about 10 minutes and maintained constant throughout the entire polymerization reaction period. After the established reaction time has passed, essentially all of the unreacted monomer(s) is/are eliminated by degassing at 60 °C at essentially atmospheric pressure.

In the second stage, the polymer product (a) of the first stage, after taking a sample for the various analyses, is brought to the established temperature for the second stage. Propylene and ethylene are then fed into the autoclave at the ratio and in the quantities established in order to achieve the pressure and the gas phase composition desired. During the polymerization the pressure and gas phase composition are maintained by feeding the propylene and ethylene mixture established by way of instruments which regulate or measure or both regulate and measure the flow rate. The length of the feed varied according to the catalyst system employed and the amount of components b) and c) desired in the particular heterophasic olefin polymer product.

At the end of the second stage polymerization reaction the powder is discharged, stabilized and then oven dried under a nitrogen stream at 60 °C.

The ingredients and relative operating conditions are set forth in Table IA and the tests results are set forth in Table IB.

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Table 1A

	Examples	1	2	3
5	1st Phase			
	Temperature, °C	70	70	70
	Pressure, atm.	31	31	31
	Time, min.	30	20	30
10	H ₂ in gas phase, mol %	0.58	0.10	0.30
	Ethylene in gas phase, mol %	1.45	2.60	2.50
	Ethylene in pol., wt. %	3.0	4.3	4.1
	Intrinsic Visc., dl/g	2.18	3.09	2.31
	Xylene Sol. (S _a), wt. %	9.4	9.0	10.7
15	Ethylene in Xylene Sol. (C _a), wt. %	11	16	17
	Intrinsic Visc. Xylene Sol.(I.V. _a), dl/g	1.15	1.39	1.19
	2nd Phase			
20	Temperature, °C	50	50	50
	Pressure, atm.	11.3	11.5	11.3
	Time, min.	335	500	250
	H ₂ in gas phase, mol %	2.23	3.0	2.05
	Ethylene in gas phase, mol %	15.9	16.9	22.54

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Table 1B

	Examples	1	2	3
	Final Product			
5	Yield, Kg Pol/g Cat	11	16.3	9.9
10	Comonomer, wt. %	24.6	22.7	29.0
	Bipolymer (b) + (c), wt. %	70	67	71.8
	Intrinsic Visc., dl/g	2.05	2.3	2.34
15	Xyl. Sol. (S _f), wt. %	63.4	60.5	63.5
	Ethylene Xyl. Sol., wt. % (C _f)	30.2	27.0	34.8
	Intrinsic Vis. Xyl. Sol. I.V. _{Sf} , dl/g	1.83	2.02	2.12
20	Fraction (b), wt. %	9.45	9.37	11.34
	Fraction (c), wt. %	60.55	57.63	60.46
	Ethylene frac. (b), wt. %	51.9	57.1	53.7
	Ethylene frac. (c), wt. %	31.1	27.6	35.7
25	Intrinsic Vis. frac. (c) (I.V. ^c), dl/g	1.86	2.05	2.18
	Melt Index, °C	150	147	145
	Flexural modulus, MPa	30	77	82
30	R.C.I. IZOD at -50 °C, J/m	NB ¹	NB	NB
	Shore D hardness	24	25	20
	Tension Set at 75%, %	41	28	36
	Tensile strength, MPa	13.8	15.8	15.4
	Tensile strength at yield, MPa	5.0	5.8	4.6
	Elongation at break, %	517	925	940
	Haze, %	31	34	35
	Vitreous transition ² , °C	-25(P) -75 -128	-23(P) -119 -121	-28(P) -81 -125

¹ NB = no break² (P) = main peak

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Examples 4

These examples illustrate a film material of the heterophasic olefin polymer composition and a method for preparing the same.

40 An air quenched blown film of the heterophasic olefin polymer composition of Example 2 stabilized with 0.25 parts per hundred parts of the olefin polymer composition (pph) octadecyl-3,5-bis(1,1-dimethylethyl)-4-hydroxybenzene propanoate, 0.05 pph tetrakis[methylene(3,5-di-tert-butyl-4-hydroxyhydrocinnamate)] methane, 0.06 pph Sandostab P-EPQ composition, the main component of which is tetrakis(2,4-di-tert-butylphenyl)-4-4'-biphenylene diphosphonite and 0.05 calcium stearate is prepared by charging the composition into a single screw extruder, extruding it through a circular die and blowing it into a film form with a sufficient amount of air to provide a film of 1 mil thickness using the following equipment and processing conditions:

- Screw: Compression ratio 3:1 to 4:1.
 Polyolefin barrier type L/D ratio = 24:1 to 30:1.
 50 Blow up ratio = 2.5 to 4.1.
 Die gap: 40 mils for a thickness of 0.5 to 5 mils.
 Extruder barrel profile: 380 to 430 °F going from zone 1 to zone 6.
 Adapter and die temperatures: 450 °F except upper and lower die zones which are 460 °F.
 Screw speed: 20 rpm.
 Pressure: 3000 psi.¹

The properties of the resulting film are set forth in Table 2 below.

¹ 1 psi = 6,895 10³ Pa

Table 2

	Properties	Resins		
		Example 2	HDPE ¹	LLDPE ²
5	Yield Strength (MD/CD ³),psi	1347/1126	3757/3145	1749/1739
	Break Strength (MD/CD), psi	3096/1643	3960/3216	2092/1898
10	Elongation at yield (MD/CD), %	65/40	30/7	80/17
	Elongation at break (MD/CD), %	361/417	120/350	317/425
	Elemendorf tear, (MD/CD), g/plly	No tear	10/282	350/790
15	Trouser tear, (MD/CD)	152/530	147/1026	503/758
	Haze	65.7	76.8	8.7
	Gloss	6	10	70
	Coefficient of friction (static/kinetic)	1.22/0.815	0.241/0.192	0.688/0.650
	Moisture vapor transmission rates at 100 °F & 100 relative humidity, g/100 sq. in/24 hrs. ¹	1.340	1.300	-
	Dart impact strength, ft-lb/mil ²	0.980	0.720	1.020

¹ Quantum LR 732 HDPE having a density of 0.953 g/cc³.

² Dowlex 2045 LLDPE containing hexene-1 and having a density of 0.920 g/cc³.

³ MD/CD = machine direction/cross direction.

¹ 1 sq. in = 6,4516•10⁻⁴ m²

² 1 ft = 0,3048 m

¹ lb = 0,4536 kg

¹ mil = 0,111 den

From the above Table one can see that the olefin polymer composition of this invention provides an air quenched blown film with improved elongation at break and Elemendorf tear properties and a better balance of properties. Further, polymer materials, the major portion of which comprise propylene units, generally do not run well, if at all, on air quenched blown film equipment.

Example 5

This example illustrates an cast film material of the heterophasic olefin polymer composition, of a coextruded film of the heterophasic olefin polymer composition and a random copolymer of propylene and ethylene and a method for preparing same.

A cast film of the heterophasic olefin polymer composition of Example 2 stabilized as set forth in Example 4 is prepared by charging the composition into an extruder, extruding it through a flat film die and quenching onto a chill roll to produce a film of 1 mil thickness using the following equipment and processing conditions:

Screw design:	Compression ratio: 4:1 to 3:1 Feed zone depth: 0.435 to 0.490" (3.5" extruder with 3.5:1 compression ratio) Metering zone depth: 0.125 to 0.140" for 3.5" extruder
Die:	Convention center-fed coathanger manifold.
Extruder operating conditions:	
Melt temperature:	430-500 °F
Extruder Barrel:	350-420 °F going from zone 1 to zone 6.
Adapter and die temperatures:	420 °F

A coextruded film 1.25 mils thick comprising a film layer of the heterophasic olefin polymer composition of Example 2 stabilized as set forth in Example 4 as the center layer, and two film layers of Pro-fax SA 861 propylene-ethylene random copolymer with an ethylene content of 3.0%, one layer of said copolymer on each surface of the center layer is prepared by the cast film technique.

The properties of the resulting film are set forth in Table 3 below.

Table 3

Properties	Resins		
	Ex. 2	Coex ¹	P-E Co ²
Yield Strength (MD/CD ³), psi	1185/865	1095/817	2523/2452
Break Strength (MD/CD), psi	3899/1708	3637/1693	4120/3363
Elongation at yield (MD/CD), %	34/19	32/22	15/10
Elongation at break (MD/CD), %	>450/>550	>530/>550	522/584
Elemendorf tear, (MD/CD), g/ply	192/No Tear	657/544	49/102
Haze	49.0	5.2	2.3
Gloss	20.0	57.0	78.1

¹ Propylene-ethylene random copolymer/Heterophasic olefin polymer composition of Ex. 2/Propylene-ethylene random copolymer.

² Pro-fax 861 propylene-ethylene random copolymer with an ethylene content of 3.0%.

³ MD/CD = machine direction/cross direction.

From the above Table one can see that the olefin polymer composition of this invention provides an cast film and a coextruded cast filme with improved elongation at yield and Elemendorf tear properties.

Various types of film materials of conventional thickness and thin films less than 20 mils thick to as thin as about 0.5 mils can be prepared using the heterophasic olefin polymer composition described herein as well as heavy film materials, typically referred to as sheets, from 20 to 100 mils thick. For example, it can be used to prepare cast films, uniaxially and biaxially oriented films and extruded or calendered sheets. In addition, a layer comprising the heterophasic olefin polymer composition can be applied to, e.g. by lamination or coextrusion techniques, at least one surface of a thermoplastic film material or a metallic sheet or foil substrate. Typical thermoplastic materials include crystalline homopolymers of a C₂-10 alpha-olefin monomer, such as propylene or ethylene, or copolymers of propylene with ethylene or with a C₄-10 alpha-olefin monomers or of propylene with both ethylene and a C₄-10 alpha-olefin monomers, provided that, when the comonomer is ethylene, the maximum polymerized ethylene content is about 10%, preferably about 4%, and when the comonomer is a C₄-10 olefin, the maximum polymerized content thereof is about 20%, preferably about 16%, and when both ethylene and an alpha-olefin are used the maximum polymerized content of both is 30%, preferably 20%, as well as polyesters, polyamides, ethylene-vinyl alcohol copolymers and ethylene-vinyl acetate copolymers. Aluminum is a suitable metallic substrate.

In addition, film materials can be prepared from blends of from about 5 to 45% of the heterophasic olefin polymer composition described herein with from about 95 to 55% of a crystalline homopolymer of a C₂-10 alpha-olefin monomer or copolymer of propylene with ethylene or with a C₄-10 alpha-olefin monomer or of propylene, ethylene and a C₄-10 alpha-olefin monomer, said copolymer having the maximum polymerized content of ethylene or alpha-olefin or both as described in the preceding paragraph. Preferably the amount of the heterophasic olefin polymer composition present in such blends is from 10 to 30%.

The olefin polymer composition of this invention is such that one can achieve the proper balance of properties in the resulting product when a film layer of same is the material applied to at least one surface of another thermoplastic material or to a metallic substrate and when it is blended with another thermoplastic material and the blend is used to make film materials.

Other features, advantages and embodiments of the invention disclosed herein will be readily apparent to those exercising ordinary skill after reading the foregoing disclosures. In this regard, while specific embodiments of the invention have been described in considerable detail, variations and modifications of these embodiments can be effected without departing from the spirit and scope of the invention as described and claimed.

Claims

1. A film or sheet material comprising an olefin polymer composition which is comprised of
 - (a) from about 10 to 50 parts of a propylene homopolymer having an isotactic index greater than 80, or a copolymer selected from the group consisting of (i) propylene and ethylene, (ii) propylene, ethylene and a CH₂ = CHR alpha-olefin, where R is a C₂-8 straight or branched alkyl, and (iii) propylene and an alpha-olefin as defined in (ii), said copolymer containing over 80% propylene and

- having an isotactic index greater than 80;
- (b) from about 5 to 20 parts of a semi-crystalline, essentially linear copolymer fraction having a crystallinity of about 20 to 60% wherein the copolymer is selected from the group consisting of (i) ethylene and propylene containing over 55% ethylene, (ii) ethylene, propylene, and an alpha-olefin as defined in (a) (ii) containing from 1 to 10% of the alpha-olefin and over 55% of both ethylene and alpha-olefin, and (iii) ethylene and an alpha-olefin as defined in (a) (ii) containing over 55% of said alpha-olefin, which copolymer is insoluble in xylene at room or ambient temperature; and
- (c) from about 40 to 80 parts of a copolymer fraction wherein the copolymer is selected from the group consisting of (i) ethylene and propylene containing from 20% to less than 40% ethylene, (ii) ethylene, propylene, and an alpha-olefin as defined in (a) (ii) wherein the alpha-olefin is present in an amount of 1 to 10% and the amount of ethylene and alpha-olefin present is from 20% to less than 40%, and (iii) ethylene and an alpha-olefin as defined in (a) (ii) containing from 20% to less than 40% of said alpha-olefin, and optionally with 0.5 to 10 % of a diene, said copolymer fraction being soluble in xylene at ambient temperature, and having an intrinsic viscosity of from 1.5 to 4.0 dl/g;
- with the total of the (b) and (c) fractions, based on the total olefin polymer composition, being from about 50% to 90%, and the weight ratio of (b)/(c) being less than 0.4.
2. The material of claim 1 wherein (a) is a copolymer of propylene and ethylene or a copolymer of propylene and butene-1.
 3. The material of claim 1 or 2 wherein (c) is a copolymer of propylene and ethylene or a terpolymer.
 4. The material of claims 1-3 wherein the total content of copolymerized ethylene is from 15 % to 35 % by weight.
 5. A film or sheet article comprising a base film or sheet of a crystalline homopolymer of a C₂₋₁₀ alpha-olefin monomer or of a copolymer selected from the group consisting of (i) propylene with ethylene, (ii) propylene with ethylene and a C₄₋₁₀ alpha-olefin monomer, and (iii) propylene with a C₄₋₁₀ alpha-olefin monomer, provided that, when the comonomer is ethylene, the maximum polymerized ethylene content is about 10 %, when the comonomer is a C₄₋₁₀ olefin, the maximum polymerized content thereof is about 20 %, and when both ethylene and a C₄₋₁₀ olefin are used the maximum polymerized content is about 30 %; and applied to at least one surface of the base film or sheet, a layer of the material of claims 1-3.
 6. A film or sheet article comprising a base film or sheet of a metallic substrate having applied to at least one surface thereof a layer of the material of claims 1-3.
 7. A film or sheet material comprising a blend of
 - (1) from about 5 to 45 % an olefin polymer composition which is comprised of
 - (a) from about 10 to 50 parts of a propylene homopolymer having an isotactic index greater than 80, or a copolymer selected from the group consisting of (i) propylene and ethylene, (ii) propylene, ethylene and a CH₂ = CHR alpha-olefin, where R is a C₂₋₈ straight or branched alkyl, and (iii) propylene and an alpha-olefin as defined in (ii), said copolymer containing over 80% propylene and having an isotactic index greater than 80;
 - (b) from about 5 to 20 parts of a semi-crystalline, essentially linear copolymer fraction having a crystallinity of about 20 to 60% wherein the copolymer is selected from the group consisting of (i) ethylene and propylene containing over 55% ethylene, (ii) ethylene, propylene, and an alpha-olefin as defined in (a) (ii) containing from 1 to 10% of the alpha-olefin and over 55% of both ethylene and alpha-olefin, and (iii) ethylene and an alpha-olefin as defined in (a) (ii) containing over 55% of said alpha-olefin, which copolymer is insoluble in xylene at room or ambient temperature; and
 - (c) from about 40 to 80 parts of a copolymer fraction wherein the copolymer is selected from the group consisting of (i) ethylene and propylene containing from 20% to less than 40% ethylene, (ii) ethylene, propylene, and an alpha-olefin as defined in (a) (ii) wherein the alpha-olefin is present in an amount of 1 to 10% and the amount of ethylene and alpha-olefin present is from 20% to less than 40%, and (iii) ethylene and an alpha-olefin as defined in (a) (ii) containing from 20% to less than 40% of said alpha-olefin, and optionally with 0.5 to 10 % of a diene, said copolymer fraction being soluble in xylene at ambient temperature, and having an intrinsic viscosity of from 1.5 to 4.0 dl/g;

viscosity of from 1.5 to 4.0 dl/g;
 with the total of the (b) and (c) fractions, based on the total olefin polymer composition, being from about 50% to 90%, and the weight ratio of (b)/(c) being less than 0.4; and
 5 (2) from about 95 to 55% of a crystalline homopolymer of a C₂-10 alpha-olefin monomer or of a copolymer selected from the group consisting of (i) propylene with ethylene, (ii) propylene with ethylene and a C₄-10 alpha-olefin monomer and (iii) propylene with a C₄-10 alpha-olefin monomer, provided that, when the comonomer is ethylene, the maximum polymerized ethylene content is about 10%, when the comonomer is a C₄-10 alpha-olefin monomer, the maximum polymerized content is about 20% and when the comonomers are ethylene and a C₄-10 alpha-olefin monomer,
 10 the maximum polymerized content of both is about 30%

- 8. The material of claim 7 wherein (1) (a) is a copolymer of propylene and ethylene, (1) (c) is a terpolymer of propylene, ethylene and butene-1.
- 15 9. The material of claim 7 wherein (1) (a) is a copolymer of propylene and butene-1, (1) (c) is a copolymer of propylene and ethylene.
- 10. The material of claim 9 wherein (1) is present in an amount from 10 to 30%.

20 Patentansprüche

1. Film- oder Folienmaterial, das eine Olefin-Polymerzusammensetzung aufweist, die
 - (a) von etwa 10 bis 50 Teile eines Propylen-Homopolymers, das einen isotaktischen Index größer als 80 aufweist, oder eines Copolymers, ausgewählt aus der Gruppe bestehend aus (i) Propylen und Ethylen, (ii) Propylen, Ethylen und einem CH₂ = CHR Alpha-Olefin, worin R ein gerades oder verzweigtes C₂-8 Alkyl ist, und (iii) Propylen und einem Alpha-Olefin, wie in (ii) definiert, wobei dieses Copolymer über 80% Propylen enthält und einen isotaktischen Index größer als 80 aufweist;
 - (b) von etwa 5 bis 20 Teile einer semikristallinen, im wesentlichen linearen Copolymer-Fraktion, die eine Kristallinität von etwa 20 bis 60% aufweist, worin das Copolymer ausgewählt ist aus der Gruppe bestehend aus (i) Ethylen und Propylen, wobei es über 55% Ethylen enthält, (ii) Ethylen, Propylen und einem Alpha-Olefin wie in (a) (ii) definiert, wobei es von 1 bis 10% das Alpha-Olefin und über 55% beides, Ethylen und Alpha-Olefin enthält, und (iii) Ethylen und einem Alpha-Olefin wie in (a) (ii) definiert, wobei es über 55% dieses Alpha-Olefin enthält, wobei dieses Copolymer in Xylool bei Raum-oder Umgebungstemperatur unlöslich ist; und
 - (c) von etwa 40 bis 80 Teile einer Copolymer-Fraktion, worin das Copolymer ausgewählt ist aus der Gruppe bestehend aus (i) Ethylen und Propylen, wobei es von 20 bis weniger als 40% Ethylen enthält, (ii) Ethylen, Propylen und einem Alpha-Olefin wie in (a) (ii) definiert, worin das Alpha-Olefin in einer Menge von 1 bis 10% vorliegt und die Menge des Ethylens und Alpha-Olefins von 20% bis weniger als 40% beträgt, und (iii) Ethylen und einem Alpha-Olefin wie in (a) (ii) definiert, wobei es von 20% bis weniger als 40% dieses Alpha-Olefin enthält, und wahlweise 0,5 bis 10% eines Dien, wobei diese Copolymerfraktion in Xylool bei Umgebungstemperatur löslich ist und eine intrinsische Viskosität von 1,5 bis 4,0 dl/g aufweist;
 enthält;

wobei die Gesamtmenge der Fraktionen (b) und (c) von etwa 50% bis 90% beträgt, basierend auf der Gesamt-Olefin-Polymerzusammensetzung, und das Gewichtsverhältnis (b)/(c) weniger als 0,4 beträgt.
2. Material gemäß Anspruch 1, worin (a) ein Copolymer aus Propylen und Ethylen oder ein Copolymer aus Propylen und Buten-1 ist.
3. Material gemäß Anspruch 1 oder 2, worin (c) ein Copolymer aus Propylen und Ethylen oder ein Terpolymer ist.
4. Material gemäß den Ansprüchen 1-3, worin der Gesamtgehalt des copolymerisierten Ethylens von 15 bis 35 Gew. % beträgt.
5. Film- oder Folienartikel, der einen Grundfilm oder eine Grundfolie aus einem kristallinen Homopolymer eines C₂-10 Alpha-Olefin-Monomers oder einem Copolymer, ausgewählt aus der Gruppe bestehend

aus (i) Propylen mit Ethylen, (ii) Propylen mit Ethylen und einem C₄-10 Alpha-Olefin-Monomer; und (iii) Propylen mit einem C₄-10 Alpha-Olefin-Monomer, aufweist, vorausgesetzt, daß wenn das Comonomer Ethylen ist, der maximal polymerisierte Ethylen Gehalt etwa 10% beträgt, wenn das Comonomer ein C₄-10 Olefin ist, der maximal polymerisierte Gehalt davon etwa 20% beträgt, und wenn beide, Ethylen und ein C₄-10 Olefin, verwendet werden, der maximal polymerisierte Gehalt etwa 30% beträgt; und auf mindestens einer Oberfläche des Grundfilms oder der Grundfolie eine Schicht des Materials der Ansprüche 1-3 aufgetragen ist.

- 5 6. Film- oder Folienartikel, der einen Grundfilm, oder eine Grundfolie aus einem metallischen Substrat aufweist, wobei auf mindestens einer Oberfläche davon eine Schicht des Materials der Ansprüche 1-3 aufgetragen ist.
- 10 7. Film- oder Folienmaterial, das ein Blend aufweist aus
 - (1) von etwa 5 bis 45% einer Olefin-Polymerzusammensetzung, die
 - 15 (a) von etwa 10 bis 50 Teile eines Propylen-Homopolymers, das einen isotaktischen Index größer als 80 aufweist, oder eines Copolymers, ausgewählt aus der Gruppe bestehend aus (i) Propylen und Ethylen, (ii) Propylen, Ethylen und einem CH₂ = CHR Alpha-Olefin, worin R ein gerades oder verzweigtes C₂-8 Alkyl ist, und (iii) Propylen und einem Alpha-Olefin, wie in (ii) definiert, wobei dieses Copolymer über 80% Propylen enthält und einen isotaktischen Index größer als 80 aufweist;
 - 20 (b) von etwa 5 bis 20 Teile einer semikristallinen, im wesentlichen linearen Copolymer-Fraktion, die eine Kristallinität von etwa 20 bis 60% aufweist, worin das Copolymer ausgewählt ist aus der Gruppe bestehend aus (i) Ethylen und Propylen, wobei es über 55% Ethylen enthält, (ii) Ethylen, Propylen und einem Alpha-Olefin wie in (a) (ii) definiert, wobei es von 1 bis 10% das Alpha-Olefin und über 55% beides, Ethylen und Alpha-Olefin enthält, und (iii) Ethylen und einem Alpha-Olefin wie in (a) (ii) definiert, wobei es über 55% dieses Alpha-Olefin enthält, wobei dieses Copolymer in Xylol bei Raum- oder Umgebungstemperatur unlöslich ist; und
 - 25 (c) von etwa 40 bis 80 Teile einer Copolymer-Fraktion, worin das Copolymer ausgewählt ist aus der Gruppe bestehend aus (i) Ethylen und Propylen, wobei es von 20 bis weniger als 40% Ethylen enthält, (ii) Ethylen, Propylen und einem Alpha-Olefin wie in (a) (ii) definiert, worin das Alpha-Olefin in einer Menge von 1 bis 10% vorliegt und die Menge des Ethylens und Alpha-Olefins von 20% bis weniger als 40% beträgt, und (iii) Ethylen und einem Alpha-Olefin wie in (a) (ii) definiert, wobei es von 20% bis weniger als 40% dieses Alpha-Olefin enthält, und wahlweise 0,5 bis 10% eines Diens, wobei diese Copolymerfraktion in Xylol bei Umgebungstemperatur löslich ist und eine intrinsische Viskosität von 1,5 bis 4,0 dl/g aufweist;
 - 30 enthält;
 - wobei die Gesamtmenge der Fraktionen (b) und (c) von etwa 50% bis 90% beträgt, basierend auf der Gesamt-Olefin-Polymerzusammensetzung, und das Gewichtsverhältnis (b)/(c) weniger als 0,4 beträgt; und
 - 35 (2) von etwa 95 bis 55% einem kristallinen Homopolymer eines C₂-10 Alpha-Olefin-Monomers oder einem Copolymer, ausgewählt aus der Gruppe bestehend aus (i) Propylen mit Ethylen, (ii) Propylen mit Ethylen und einem C₄-10 Alpha-Olefin-Monomer, und (iii) Propylen mit einem C₄-10 Alpha-Olefin-Monomer, vorausgesetzt, daß wenn das Comonomer Ethylen ist, der maximal polymerisierte Ethylengehalt etwa 10% beträgt, wenn das Comonomer ein C₄-10 Alpha-Olefin-Monomer ist, der maximal polymerisierte Gehalt etwa 20% beträgt, und wenn die Comonomere Ethylen und ein C₄-10 Alpha-Olefin-Monomer sind, der maximal polymerisierte Gehalt von beiden etwa 30% beträgt.
 - 40 8. Material gemäß Anspruch 7, worin (1) (a) ein Copolymer aus Propylen und Ethylen und (1) (c) ein Terpolymer aus Propylen, Ethylen und Buten-1 ist.
 - 45 9. Material gemäß Anspruch 7, worin (1) (a) ein Copolymer aus Propylen und Buten-1 und (1) (c) ein Copolymer aus Propylen und Ethylen ist.
 - 50 10. Material gemäß Anspruch 9, worin (1) in einer Menge von 10 bis 30% vorhanden ist.

Revendications

1. Matière pelliculaire ou en feuille comprenant une composition polymère oléfinique qui est composée de :
 - 5 (a) environ 10 à 50 parties d'un homopolymère de propylène ayant un indice isotactique supérieur à 80, ou un copolymère choisi parmi le groupe consistant en :
 - (i) du propylène et de l'éthylène;
 - (ii) du propylène, de l'éthylène et une α -oléfine $\text{CH}_2 = \text{CHR}$, où R est un alcoyle linéaire ou branché en C_{2-8} , et
 - (iii) du propylène et une α -oléfine telle que définie en (ii),
 - 10 le copolymère contenant plus de 80% de propylène et ayant un indice isotactique supérieur à 80;
 - (b) environ 5 à 20 parties d'une fraction semicristalline d'un copolymère essentiellement linéaire, ayant une cristallinité d'environ 20 à 60%, dans laquelle le copolymère est choisi parmi le groupe consistant en :
 - 15 (i) de l'éthylène et du propylène contenant plus de 55% d'éthylène;
 - (ii) de l'éthylène, du propylène et une α -oléfine telle que définie en (a)(ii), contenant de 1 à 10% de l' α -oléfine et plus de 55% d'éthylène et d' α -oléfine, et
 - (iii) de l'éthylène et une α -oléfine telle que définie en (a)(ii), contenant plus de 55% de l' α -oléfine, le copolymère est insoluble dans le xylène à température ambiante, et
 - 20 (c) environ 40 à 80 parties d'une fraction d'un copolymère, dans laquelle le copolymère est choisi parmi le groupe consistant en :
 - (i) de l'éthylène et du propylène contenant de 20% à moins de 40% d'éthylène;
 - (ii) de l'éthylène, du propylène et une α -oléfine telle que définie en (a)(ii), où l' α -oléfine est présente en une quantité de 1 à 10% et la quantité d'éthylène et d' α -oléfine présente se situe entre 20% et moins de 40%, et
 - (iii) de l'éthylène et une α -oléfine telle que définie en (a)(ii) contenant de 20% à moins de 40% de l' α -oléfine,
 et facultativement, avec 0,5 à 10% d'un diène, la fraction de copolymère étant soluble dans le xylène à température ambiante et ayant une viscosité intrinsèque de 1,5 à 4,0 dl par g; le total des fractions (b) et (c), sur base de la composition polymère oléfinique totale, étant compris entre environ 50% et 90%, et le rapport pondéral de (b)/(c) étant inférieur à 0,4.
2. Matière suivant la revendication 1, dans laquelle (a) est un copolymère de propylène et d'éthylène ou un copolymère de propylène et de butène-1.
- 35 3. Matière suivant la revendication 1 ou 2, dans laquelle (c) est un copolymère de propylène et d'éthylène ou un terpolymère.
4. Matière suivant les revendications 1 à 3, dans laquelle la teneur totale en éthylène copolymérisé se situe de 15% à 35% en poids.
- 40 5. Article pelliculaire ou en feuille comprenant une pellicule ou feuille de base, d'un homopolymère cristallin d'un monomère α -oléfinique en C_{2-10} ou d'un copolymère choisi parmi le groupe consistant en
 - (i) du propylène avec de l'éthylène;
 - (ii) du propylène avec de l'éthylène et un monomère α -oléfinique en C_{4-10} , et
 - (iii) du propylène avec un monomère α -oléfinique en C_{4-10} , pourvu que, lorsque le comonomère est de l'éthylène, la teneur maximale en éthylène copolymérisé est d'environ 10%, lorsque le comonomère est une oléfine en C_{4-10} , sa teneur maximale copolymérisée est d'environ 20% et lorsque l'éthylène et une oléfine en C_{4-10} sont utilisés, la teneur polymérisée maximale est d'environ 30%; et une couche de matière suivant les revendications 1 à 3, appliquée sur au moins une surface de la pellicule ou feuille de base.
- 55 6. Article pelliculaire ou en feuille comprenant une pellicule ou feuille de base d'un substrat métallique, sur au moins une des surfaces de laquelle on a appliqué une couche de la matière suivant les revendications 1 à 3.
7. Matière pelliculaire ou en feuille, comprenant un mélange de :

- (1) environ 5 à 45% d'une composition polymère oléfinique qui est composée de :
- (a) environ 10 à 50 parties d'un homopolymère de propylène ayant un indice isotactique supérieur à 80, ou un copolymère choisi parmi le groupe consistant en :
 - (i) du propylène et de l'éthylène;
 - (ii) du propylène, de l'éthylène et une α -oléfine $\text{CH}_2 = \text{CHR}$, où R est un alcoyle linéaire ou branché en C_{2-8} , et
 - (iii) du propylène et une α -oléfine telle que définie en (ii),
 le copolymère contenant plus de 80% de propylène et ayant un indice isotactique supérieur à 80;
 - (b) environ 5 à 20 parties d'une fraction semicristalline d'un copolymère essentiellement linéaire, ayant une cristallinité d'environ 20 à 60%, dans laquelle le copolymère est choisi parmi le groupe consistant en :
 - (i) de l'éthylène et du propylène contenant plus de 55% d'éthylène;
 - (ii) de l'éthylène, du propylène et une α -oléfine telle que définie en (a)(ii), contenant de 1 à 10% de l' α -oléfine et plus de 55% d'éthylène et d' α -oléfine, et
 - (iii) de l'éthylène et une α -oléfine telle que définie en (a)(ii), contenant plus de 55% de l' α -oléfine, le copolymère est insoluble dans le xylène à température ambiante, et
 - (c) environ 40 à 80 parties d'une fraction d'un copolymère, dans laquelle le copolymère est choisi parmi le groupe consistant en :
 - (i) de l'éthylène et du propylène contenant de 20% à moins de 40% d'éthylène;
 - (ii) de l'éthylène, du propylène et une α -oléfine telle que définie en (a)(ii), où l' α -oléfine est présente en une quantité de 1 à 10% et la quantité d'éthylène et d' α -oléfine présente se situe entre 20% et moins de 40%, et
 - (iii) de l'éthylène et une α -oléfine telle que définie en (a)(ii) contenant de 20% à moins de 40% de l' α -oléfine, et facultativement, avec 0,5 à 10% d'un diène, la fraction de copolymère étant soluble dans le xylène à température ambiante et ayant une viscosité intrinsèque de 1,5 à 4,0 dl par g; le total des fractions (b) et (c), sur base de la composition polymère oléfinique totale, étant compris entre environ 50% et 90%, et le rapport pondéral de (b)/(c) étant inférieur à 0,4, et
- (2) environ 95 à 55% d'un homopolymère cristallin d'un monomère α -oléfinique en C_{2-10} ou d'un copolymère choisi parmi le groupe consistant en :
- (i) du propylène avec de l'éthylène;
 - (ii) du propylène avec de l'éthylène et un monomère α -oléfinique en C_{4-10} , et
 - (iii) du propylène avec un monomère α -oléfinique en C_{4-10} , pourvu que, lorsque le comonomère est l'éthylène, la teneur maximale en éthylène polymérisé est d'environ 10%, lorsque le comonomère est un monomère α -oléfinique en C_{4-10} , la teneur polymérisée maximale est d'environ 20% et lorsque les comonomères sont l'éthylène et un monomère α -oléfinique en C_{4-10} , la teneur polymérisée maximale de ceux-ci est d'environ 30%.
8. Matière suivant la revendication 7, dans laquelle (1)(a) est un copolymère de propylène et d'éthylène et (1)(c) est un terpolymère de propylène, d'éthylène et de butène-1.
9. Matière suivant la revendication 7, dans laquelle (1)(a) est un copolymère de propylène et de butène-1 et (1)(c) est un copolymère de propylène et d'éthylène.
10. Matière suivant la revendication 9, dans laquelle (1) est présent en une quantité de 10 à 30%.